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Preliminary Draft "Biomedical Considerations
Related To Inhaled Alpha-Emitting
Isotopes" Los Alamos Foundation

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V. RADIATION PROTECTION STANDARDS FOR ^{239}Pu

A. Historical Development

Recent reviews of radiation protection standards for ^{239}Pu have been published by Langham (1972) and Healy (1970). Both of these reviews trace the current standards back to primarily human data derived from individuals containing quantities of ^{226}Ra or those exposed to external X- or gamma irradiation. In 1943, during the early days of the Manhattan Project, there were only three tolerance values available: 0.1 R/day for external X- or gamma irradiation, 0.1 μg body burden for ^{226}Ra and radon air concentrations of 1×10^{-11} Ci/liter. Early biological experimentation demonstrated the bone-seeking nature of ^{239}Pu when it was absorbed into body fluids in some forms. Because the work of Evans *et al.* (1943) on humans containing body burdens of ^{226}Ra was available in this time period, it was natural that comparisons of the expected toxicity of ^{239}Pu in man be made with that observed for ^{226}Ra in humans. During the early days of the Manhattan Project, the permissible body burden of ^{239}Pu in 1944 was initially taken as 0.3 μCi but was reduced to 0.06 or 0.03 μCi in different laboratories in 1945.

With the availability of comparative toxicity data for ^{226}Ra and ^{239}Pu in rodents, the permissible body burden for ^{239}Pu was reviewed at the Tripartite Permissible Doses Conference held at Chalk River, Canada in September 1949. At this time, a permissible body burden of 0.006 μCi was agreed upon. Shortly thereafter, the value was revised to 0.04 μCi based on an analysis by Dr. Austin Brues that accounted for (a) a 15:1 toxicity ratio for ^{239}Pu relative to ^{226}Ra based on injected doses, (b) 75% ^{239}Pu retention versus 25% ^{226}Ra retention, and (c) 15-20% Rn retention in the rodent compared with ~50% for man. From these factors, $1 \mu\text{Ci } ^{226}\text{Ra} = \frac{1}{15} \times \frac{0.75}{0.25} \left(\frac{4.8 + 0.5}{4.8 + 0.15} \frac{(5.5 + 6.0 + 7.7)}{(5.5 + 6.0 + 7.7)} \right) = \sim 0.4 \mu\text{Ci}$. Since the permissible body burden for ^{226}Ra was, and still is, 0.1 μCi , the resulting value for ^{239}Pu was 0.04 μCi . This permissible body burden for ^{239}Pu based on the skeleton as the critical organ has been used by the ICRP since 1950, National Bureau of Standards (1951) and the NCRP since 1953, National Bureau of Standards (1953).

Cognizance of the differences that in vivo solubility of an inhaled material can play in the subsequent tissue distribution was made by the NCRP in 1951 when they tabulated the maximum permissible body burden, MPBB, for ^{239}Pu in both soluble and insoluble forms, National Bureau of Standards (1953). For insoluble ^{239}Pu , with lung as the critical organ, the MPBB was 0.008 μCi , a value

... calculation based on a permissible weekly mean organ dose of 0.3 rep/... where the RBE for alpha particles was apparently taken as 20. The ICRP... shed a value of 0.02 derived similarly in 1955 using an RBE of 10. Subsequent... lications by the NCRP and ICRP have listed maximum permissible air concentra-... ons, MPC_a for insoluble forms of ²³⁹Pu derived from calculations in which the annual dose to lung was limited to 15 rem. Although corresponding MPBB values have not been listed, an annual average dose of this magnitude to lung would be associated with a lung burden of 0.016 μCi. Thus, the recommended value for insoluble ²³⁹Pu in the lung is based on irradiation limits to a critical tissue, a concept that traces back originally to human experience with external irradiation.

B. Current Standards

The 1959 recommendations of the NCRP, National Bureau of Standards (1959) and the ICRP (1960) regarding maximum permissible body burdens and associated maximum permissible concentrations in air and water for ²³⁹Pu which are the currently accepted values, are given below:

| Form | Critical Organ | Maximum Permissible Burden in Total Body (μCi) | Occupational MPC _a (μCi/cc) | |
|-----------|----------------|--|--|---------------------|
| | | | 40 hr | 168 hr |
| Soluble | Bone | 0.04 | 2×10^{-12} | 6×10^{-13} |
| Insoluble | Lung | ---- | 4×10^{-11} | 1×10^{-11} |

Current AEC standards as published in AECM 0524, USAEC (1968) and 10 CFR20, USAEC (1971) for ²³⁹Pu for 40 hours per week exposures in restricted areas are the same as those recommended by NCRP and ICRP.

One of the main problems with these standards, as they now exist, is the differentiation between soluble and insoluble forms of ²³⁹Pu in the industrial situation. Recognizing that there is probably an entire spectrum of in vitro solubilities for different compounds of ²³⁹Pu, it is not clear on what basis the decision should be made regarding use of the soluble or insoluble limits. This decision is an important one because the limits now differ by a factor of 20.

As was indicated above, the permissible annual dose to lung, 15 rem, can be traced back to human experience with external irradiation. More needs to be learned of the relationship between the uniform dose produced by the external irradiation and the more localized doses to lung produced following inhalation of alpha-emitting particulates.

The standard for soluble ^{239}Pu assumes that essentially all of the internally deposited ^{239}Pu is in the skeleton. Results from animal experimentation have shown that the partition of ^{239}Pu between liver and skeleton after it reaches the bloodstream is variable and influenced by route of administration, chemical form and other factors. Thus, a standard that requires all ^{239}Pu to be in the skeleton is unrealistic. Bair and Thompson (1974) have proposed a scheme with 45% in skeleton, 45% in liver and the remaining 10% in soft tissue and excreta.

C. Potential Changes

Bair and Thompson (1974), in their recent review of the biomedical aspects of plutonium, examined the relationship of present standards to existing animal data. Using comparative data on ^{226}Ra and ^{239}Pu obtained with Beagle dogs at the University of Utah and making some assumptions regarding differences in bone surface/volume ratios and turnover of ^{239}Pu on bone surfaces for humans, they concluded that the current standard for plutonium was severalfold "less safe" for bone than the current standard for radium. Further, they compared the lung burden in man associated with an annual dose of 15 rems, 0.016 μCi , with lung burdens shown to have produced pulmonary neoplasia in dogs. On a concentration basis, an initial lung concentration of ~ 1 nCi/gm might not shorten the lifespan of the dog. This value can be compared with the permissible lung concentration in man of 0.016 nCi/gm. Because of the absence of data at lower levels of ^{239}Pu , the authors deemed the margin of safety "less than totally reassuring." They also pointed out differences that could be incurred if the tracheo-bronchial lymph nodes or the liver were considered the critical organ. They summarized their thoughts in this area by indicating that they both feel that there will be some lowering of permissible exposures to ^{239}Pu within the next few years but that the change will probably not be large.

In a recent review, Dolphin (1972) discussed problems associated with selection of a critical organ for control of the exposure of people to plutonium. He reviewed the animal data and the observation of long-term effects in lung, lymph tissue associated with lung, bone and liver and concluded that the choice was not clear-cut, particularly considering the differences between the high body burdens required to produce observable changes in laboratory animals compared with the low levels and anticipated minimal response expected in a human population. He then used a revised version of a lung model derived by the ICRP Task Group on Lung Dynamics (1966) that accounted for different retention times for different compounds in the lung and its associated lymph nodes, and also took account of differences in aerosol particle size distribution. For a particle size distribution with an activity median aerodynamic

diameter of 1 μm , he concluded that a MPC_a of $1 \times 10^{-11} \mu\text{Ci/cc}$ would give adequate protection for both Class W and Class Y compounds. This value would be 4 times more restrictive than the current MPC_a for insoluble ^{239}Pu but 5 times less restrictive than the current MPC_a for soluble forms.

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